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13. ABSTRACT *(Maximum 200 words)*

This is the final report on an Army Research Office grant supporting a fellowship. Two fellows were supported by the grant. One of the fellows developed a new technique to study gas phase molecules at liquid helium temperatures. The second used time-resolved double resonance techniques to determine the state dependence and cross section of many rotational and vibrational processes in methyl halides.

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NEAR MILLIMETER WAVES AND MICROELECTRONICS

FINAL REPORT

FRANK C. DE LUCIA

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U.S. ARMY RESEARCH OFFICE

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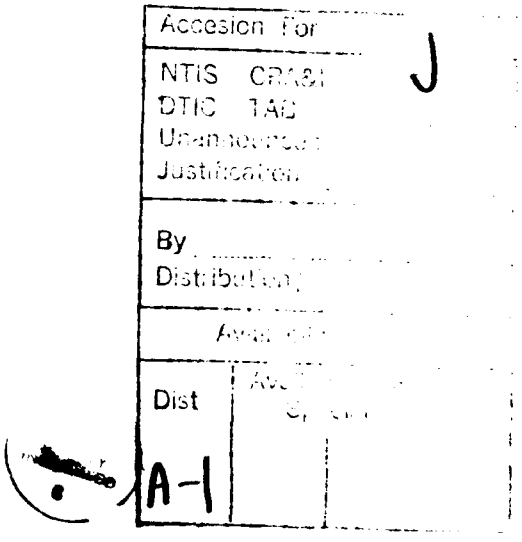

Statement of the Problem: The purpose of this grant was to support an Army Research Office Fellow. The fellows supported were Daniel R. Willey and Henry O. Everitt, III. Mr. Willey developed a new technique to study gas phase molecules at liquid helium temperatures. His work significantly improved the link between the theoretical descriptions of molecules and experimental investigations.

Mr. Everitt used time-resolved double resonance techniques to determine the state dependence and cross section of many rotational and vibrational state-changing processes in methyl halides. His work greatly furthered fundamental understanding of optically pumped far infrared lasers.

Summary of the most important results:

Low temperature spectroscopy: Sample gas molecules were cooled by collisions with helium atoms in a cell immersed in a liquid helium bath. At liquid helium temperatures only the lowest lying rotational states have significant population. This greatly simplifies theoretical calculations and makes possible a more direct link between experiment and theory. In addition, working at these low temperatures confers the advantages of narrow linewidths, large absorption coefficients and the simplification of complex spectra on the spectroscopic investigations.

Low temperature (1.8 - 4.2 K) pressure broadening by helium was measured for CO, CH_3F, H_2S, NO, DCL, CH_2F_2, and HDO. The pressure broadening cross sections ranged from 79.2 Angstroms squared to 8.1 Angstroms squared. Although the cross sections for CO and CH_3F showed an initial rise below 4K, at the lowest temperatures all of the cross sections appeared to be declining. These investigations appear to indirectly confirm resonant structure predicted by theory in the cross section of CO-He and to indicate the possible existence of similar resonances in CH_3F-He, CH_2F_2-He, and DCL-He.

Rotational - Vibrational Energy Transfer: Time resolved infrared - millimeter wave double resonance techniques were used to study rotational and vibrational state processes in two isotopes of methyl fluoride and in methyl chloride. Processes which populate target states thermally as well as those which populate the target states in a rotationally nonthermal manner were observed. Nonthermal interactions of both the dipole and higher - order multipole moments of colliding molecules were studied. The dipole interactions were dominant.

In addition to the well known thermal processes of wall collisions and vibrational state-changing collisions, a vibrational quantum number swapping mechanism was discovered. Active in both halides studied, this process thermalized rotational states and symmetry types. A symmetry preserving process with a very large cross section was also discovered. This process rotationally "thermalizes" molecules. The experimentally measured cross sections of these two thermal processes have different temperature dependencies, implying different physical origins. The composite effect of all collisional processes agreed with predictions of pressure broadening studies in methyl fluoride.

A numerical simulation was developed in order to model collisional energy

transfer. In addition to facilitating the measurement of cross sections, the simulation predicts the behavior of optically pumped far infrared lasers. Experimental studies of the pressure dependent and pump power dependent behavior of optically pumped far infrared lasers contradicted previously accepted theories. However, the simulation, using only physically meaningful and redundantly verified parameters, accurately characterized the action of these lasers in all regimes.

List of Publications:

"Very Low Temperature Spectroscopy: The Pressure Broadening Coefficients for CH_3F between 4.2 and 1.9 K," Daniel R. Willey, Richard L. Crownover, D. N. Bitner, and Frank C. De Lucia, J. Chem. Phys. 89, 6147-6149 (1988).

"Very Low Temperature Spectroscopy: The Pressure Broadening Coefficients for CO-He between 4.3 and 1.7 K," Daniel R. Willey, Richard L. Crownover, D. N. Bitner, and Frank C. De Lucia, J. Chem. Phys. 89, 1923-1928 (1988).

"Collisional Cooling of the NO-He System: The Pressure Broadening Cross Sections between 4.3 and 1.8 K," Daniel R. Willey, D. N. Bitner, and Frank C. De Lucia, Mol. Phys. 67, 455-463 (1989).

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"Pressure Broadening Cross Sections for the H_2S-He System in the Temperature Region Between 4.3 and 1.8 K," Daniel R. Willey, D. N. Bitner, and Frank C. De Lucia, J. Mol. Spectrosc. 134, 240-242 (1989).

"A Time-resolved Study of Rotational Energy Transfer into A and E Symmetry Species of $^{13}CH_3F$," Henry O. Everitt and Frank C. De Lucia, J. Chem. Phys. 90, 3520-3527 (1989).

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"Frequency Stability and Reproducibility of Optically Pumped Far-Infrared Lasers," Richard L. Crownover, Henry O. Everitt, Frank C. De Lucia and David D. Skatrud, Appl. Phys. Lett. 57 (27), 2882-2884 (1990).

Participating Scientific Personnel:

Frank C. De Lucia

David D. Skatrud
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Daniel R. Willey, Ph.D. awarded 1989
Henry O. Everitt, III, Ph.D. awarded 1990

Report of Inventions: None

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Daniel R. Willey, "The Collisional Cooling Technique: Gas Phase Spectroscopy Below 5 K," Ph.D. dissertation (unpublished), Duke University, Durham, NC (1989).

Henry O. Everitt, III, "Collisional Energy Transfer in Methyl Halides," Ph.D. dissertation (unpublished), Duke University, Durham, NC (1990).